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October 3, 2006

M&C+SNA 2007
Monterey, CA, United States
April 15, 2007 through April 19, 2007

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ASYMPTOTIC ANALYSIS OF TIME-DEPENDENT NEUTRON TRANSPORT COUPLED WITH ISOTOPIC DEPLETION AND RADIOACTIVE DECAY

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ABSTRACT

We describe an asymptotic analysis of the coupled nonlinear system of equations describing time-dependent three-dimensional monoenergetic neutron transport and isotopic depletion and radioactive decay. The classic asymptotic diffusion scaling of Larsen and Keller [1], along with a consistent small scaling of the terms describing the radioactive decay of isotopes, is applied to this coupled nonlinear system of equations in a medium of specified initial isotopic composition. The analysis demonstrates that to leading order the neutron transport equation limits to the standard time-dependent neutron diffusion equation with macroscopic cross sections whose number densities are determined by the standard system of ordinary differential equations, the so-called Bateman equations, describing the temporal evolution of the nuclide number densities.

Key Words: Neutron transport, asymptotic analysis, isotopic depletion, radioactive decay.

1. INTRODUCTION

The accurate modelling of isotopic depletion and decay in a nuclear reactor core is important for several reasons. Isotopic depletion significantly affects the operating life of the reactor core fuel, the expected energy production over the life of the reactor core fuel, the core power distribution, and the stability and control of the reactor core [2, 3]. The coupled computation of the time-dependent neutron angular flux and the isotopic number densities in a nuclear reactor is a mathematically nonlinear problem: the time-dependent neutron angular flux depends on the time-dependent isotopic composition and vice versa. As a result of the computational expense of solving this coupled neutron angular flux and isotopic composition system using a neutron transport-based calculation (e.g. a discrete ordinates or spherical harmonics approximation), the neutron diffusion approximation is often used as a more computationally efficient alternative.

Asymptotic analysis methods have been used to theoretically investigate neutron transport problems for over thirty-five years. The earliest work referenced in the literature is the unpublished work of Matkowsky [4] and Habetler and Matkowsky [5], eventually leading to the publication of Ref. [6]. Larsen and Keller [1], Larsen [7], and others continued and expanded this work during the 1970's as detailed in the review article by Larsen [10]. The main focus of this early asymptotic analysis was as an approximate theoretical method for obtaining solutions of neutron transport problems. This early work also theoretically clarified the conditions under which neutron transport theory limits to neutron diffusion theory. In the intervening decades, the applications of asymptotic analysis to neutron transport problems have become more diverse. Asymptotic analysis has been employed to derive the planar geometry P_N approximation

from planar geometry transport theory [11] and the multidimensional simplified P_N approximation [12, 13] from multidimensional transport theory. Asymptotic methods have also been used to theoretically investigate the behavior of spatial discretization schemes for the discrete ordinates equations [see Ref. [14] and the references contained therein]. As an aside, we note that asymptotic methods have also been successfully applied to the analysis of radiative transfer and charged particle transport problems.

While the application of asymptotic methods to neutron transport problems has served to clarify the connection between neutron transport theory and neutron diffusion theory, to our knowledge the equations describing the temporal evolution of the isotopic number densities have never been directly included in these asymptotic analyses. In previous asymptotic analyses of time-dependent neutron transport, the material properties of the background medium (i.e. macroscopic total, absorption, and scattering cross sections) have either been assumed independent of time [1, 6, 8] or have been postulated to be slowly varying functions of time [9]. The time dependence of these material properties is in reality explicitly determined by the time dependence of the isotopic number densities present in the background medium. Given that the neutron angular flux and the isotopic number densities interact in a nonlinear manner, directly including these effects in a theoretical analysis of the system seems prudent. The work we describe in this paper is a first step towards directly including the equations describing isotopic depletion and decay in an asymptotic analysis of neutron transport theory. Further strengthening the theoretical foundation of the neutron diffusion approximation coupled with the equations describing isotopic depletion and radioactive decay is important given the fundamental and ubiquitous role this model plays in nuclear reactor design and analysis.

In this paper, we consider time-dependent monoenergetic neutron transport in a three-dimensional medium. Different than previous work, however, we consider a background medium that is initially composed of a specified isotopic composition. For the mathematical statement of the physical problem to be fully specified, the neutron transport equation must be coupled to a system of first-order differential equations describing the temporal evolution of the isotopic number densities. Both the neutron transport equation and the so-called Bateman equations describing the depletion and radioactive decay of the isotopes present are nonlinear. Using the classic asymptotic diffusion limit scaling [1, 14], we demonstrate that neutron transport theory for this system asymptotically limits to the standard time-dependent neutron diffusion approximation with macroscopic cross sections whose isotopic number densities are determined by the standard system of ordinary differential equation describing the temporal evolution of the nuclide number densities.

The remainder of this paper is organized as follows. In Section 2, we describe the mathematical specification of the neutron transport problem to be solved. We then recast the problem into a form that is amenable to the application of the classic asymptotic diffusion scaling. Finally, we outline the asymptotic analysis of the neutron transport problem. We conclude with a brief discussion in Section 3.

2. ASYMPTOTIC ANALYSIS

We consider the following time-dependent monoenergetic neutron transport problem defined in a three-dimensional isotropically-scattering medium in the spatial domain D :

$$\frac{1}{v} \frac{\partial}{\partial t} \psi(\underline{r}, \underline{\Omega}, t) + \underline{\Omega} \cdot \underline{\nabla} \psi(\underline{r}, \underline{\Omega}, t) + \Sigma_t(\underline{r}, t) \psi(\underline{r}, \underline{\Omega}, t) = \frac{1}{4\pi} \Sigma_s(\underline{r}, t) \int_{4\pi} \psi(\underline{r}, \underline{\Omega}', t) d\Omega' + \frac{1}{4\pi} Q(\underline{r}, t) \quad , \quad \underline{r} \in D \quad , \quad t > 0 \quad , \quad (1)$$

$$\psi(\underline{r}, \underline{\Omega}, t) = f(\underline{r}, \underline{\Omega}, t) \quad , \quad \underline{r} \in \partial D \quad , \quad \underline{\Omega} \cdot \underline{n} < 0 \quad , \quad t > 0 \quad , \quad (2)$$

$$\psi(\underline{r}, \underline{\Omega}, 0) = g(\underline{r}, \underline{\Omega}) \quad , \quad \underline{r} \in D \quad . \quad (3)$$

Here we have used standard neutronics notation [2, 15]: \underline{r} [cm] is the three-dimensional Cartesian spatial variable; $\underline{\Omega}$ is a unit vector describing the direction of neutron flight; t [s] is the time variable; v [cm/s] is the neutron speed of flight; $\psi(\underline{r}, \underline{\Omega}, t)$ [neutrons/cm² – steradian – s] is the angular flux (neutron density [neutrons/cm³ – steradian] multiplied by the neutron speed of flight v [cm/s]) of neutrons at position \underline{r} moving with speed v in direction $\underline{\Omega}$ at time t ; $\Sigma_t(\underline{r}, t)$ [cm⁻¹] is the macroscopic total cross section at position \underline{r} and time t describing the probability per unit path length of a neutron interacting with the background medium; $\Sigma_s(\underline{r}, t)$ [cm⁻¹] is the macroscopic isotropic scattering cross section describing the probability per unit path length of a neutron at position \underline{r} undergoing a scattering reaction; and $Q(\underline{r}, t)$ [neutrons/cm³ – s] is an external volumetric source of neutrons at position \underline{r} and time t . Here $f(\underline{r}, \underline{\Omega}, t)$ [neutrons/cm² – steradian – s] is a prescribed incident angular flux on the boundary of the spatial domain ∂D , and \underline{n} is a unit outward normal on the boundary of the system. Finally, $g(\underline{r}, \underline{\Omega})$ [neutrons/cm² – steradian – s] is a prescribed angular flux initial condition.

We assume that the background medium is initially composed of a known isotopic composition. We also assume that I distinct nuclide species exist that are either present initially or that can be created by isotopic depletion and accretion or by radioactive decay. We denote the number density [cm⁻³] of the i^{th} nuclide at position \underline{r} and time t as $N_i(\underline{r}, t)$ and the $I \times 1$ vector function of all nuclide number densities as $\underline{N}(\underline{r}, t)$.

A neutron and an atom of a specific nuclide can undergo either a scattering reaction or an absorption reaction. A scattering reaction changes the direction of the neutron exiting the collision but does not alter the isotope of the background nuclide. We denote the microscopic scattering cross section for nuclide i as σ_s^i [cm²] and the $I \times 1$ vector of microscopic scattering cross sections for all nuclide species as $\underline{\sigma}_s$. An absorption reaction can be one of several reaction types such as (n, γ) capture, (n,p), (n, α), etc. An absorption reaction results in the removal of the incident neutron, a conversion of the nucleus undergoing the reaction to another isotope, and the accretion of an additional atom of the created isotope (we assume that all reaction residual products are deposited locally and instantaneously, e.g. a helium-4 atom in the case of an (n, α) reaction). We denote the microscopic absorption cross section for nuclide i , σ_a^i [cm²], as the sum of all capture reactions and the $I \times 1$ vector of microscopic absorption cross sections for all nuclide species as $\underline{\sigma}_a$. We assume that microscopic cross sections are independent of time, i.e. no neutron spectrum or self-shielding effects [2] are present. The macroscopic cross section for a general reaction type x ($x = s, a$) can then be written in vector notation as

$$\Sigma_x(\underline{r}, t) = \underline{N}^T(\underline{r}, t) \underline{\sigma}_x \quad . \quad (4)$$

The macroscopic total cross section is the sum of the macroscopic scattering and absorption cross sections:

$$\Sigma_t(\underline{r}, t) = \Sigma_s(\underline{r}, t) + \Sigma_a(\underline{r}, t) \quad . \quad (5)$$

Using Eqs. (4) and (5), we rewrite Eq. (1) as

$$\frac{1}{v} \frac{\partial}{\partial t} \psi(\underline{r}, \underline{\Omega}, t) + \underline{\Omega} \cdot \underline{\nabla} \psi(\underline{r}, \underline{\Omega}, t) + \underline{N}^T(\underline{r}, t) \underline{\sigma}_t \psi(\underline{r}, \underline{\Omega}, t) = \frac{1}{4\pi} \underline{N}^T(\underline{r}, t) (\underline{\sigma}_t - \underline{\sigma}_a) \int_{4\pi} \psi(\underline{r}, \underline{\Omega}', t) d\underline{\Omega}' + \frac{1}{4\pi} Q(\underline{r}, t) \quad (6)$$

The equations describing the transport of neutrons, Eq. (6) and Eqs. (2) and (3), are coupled to equations describing the temporal evolution of the isotopic composition of the background medium through which the neutrons are propagating, the so-called Bateman equations. The temporal evolution of $\underline{N}(\underline{r}, t)$ is given by [2, 3]

$$\frac{\partial}{\partial t} \underline{N}(\underline{r}, t) = \underline{A} \underline{N}(\underline{r}, t) \int_{4\pi} \psi(\underline{r}, \underline{\Omega}, t) d\underline{\Omega} + \underline{\Lambda} \underline{N}(\underline{r}, t) \quad (7)$$

with the initial condition

$$\underline{N}(\underline{r}, 0) = \underline{N}_0(\underline{r}) \quad (8)$$

In Eqs. (7), the elements of the $I \times I$ absorption matrix \underline{A} are given by

$$a_{ij} = \begin{cases} -\sigma_a^j, & i = j, \\ f_{ij} \sigma_a^j, & i \neq j, \end{cases} \quad (9)$$

where f_{ij} is the fraction of neutron absorptions by nuclide species j that leads to the formation of nuclide species i . The diagonal terms of the matrix \underline{A} correspond to the depletion of the given nuclide as a result of isotopic burnup. The off-diagonal terms correspond to the production of the given nuclide as a result of isotopic depletion of other isotopes. Also in Eqs. (7), the elements of the $I \times I$ radioactive decay matrix $\underline{\Lambda}$ are given by

$$\lambda_{ij} = \begin{cases} -\lambda_j, & i = j, \\ \ell_{ij} \lambda_j, & i \neq j, \end{cases} \quad (10)$$

where λ_j is the radioactive decay constant [s^{-1}] of nuclide j , and ℓ_{ij} is the fraction of decays of nuclide species j that leads to the formation of nuclide species i . The diagonal terms of the radioactive decay matrix $\underline{\Lambda}$ correspond to the loss of the given nuclide as a result of radioactive decay. The off-diagonal terms correspond to the production of the given nuclide as a result of radioactive decay of other isotopes. The coupled computation of the time-dependent neutron angular flux from Eq. (6) and the isotopic number densities from Eqs. (7) is a nonlinear mathematical problem.

To proceed with the asymptotic analysis, we first decompose the initial value of a macroscopic cross section for a general reaction type x , $\Sigma_x(\underline{r}, 0)$, into “typical” number density and microscopic cross section values that we denote as $\langle N(\underline{r}) \rangle$ [cm^{-3}] and $\langle \sigma_x \rangle$ [cm^2], respectively, such that

$\Sigma_x(\underline{r}, 0) = \langle N(\underline{r}) \rangle \langle \sigma_x \rangle$. We next define a dimensionless number density for nuclide species i as $N_i(\underline{r}, t) = N_i(\underline{r}, t) / \langle N(\underline{r}) \rangle$ and a dimensionless microscopic cross section for nuclide i and reaction type x as $\hat{\sigma}_x^i = \sigma_x^i / \langle \sigma_x \rangle$. We then rewrite the macroscopic cross section of type x as

$$\Sigma_x(\underline{r}, t) = \underline{\hat{N}}^T(\underline{r}, t) \underline{\hat{\sigma}}_x \Sigma_x(\underline{r}, 0) \quad (11)$$

Using Eq. (11), we rewrite Eq. (6) as

$$\frac{1}{v} \frac{\partial}{\partial t} \psi(\underline{r}, \underline{\Omega}, t) + \underline{\Omega} \cdot \underline{\nabla} \psi(\underline{r}, \underline{\Omega}, t) + \underline{\hat{N}}^T(\underline{r}, t) \underline{\hat{\sigma}}_t \Sigma_t(\underline{r}, 0) \psi(\underline{r}, \underline{\Omega}, t) = \frac{1}{4\pi} \underline{\hat{N}}^T(\underline{r}, t) [\underline{\hat{\sigma}}_t \Sigma_t(\underline{r}, 0) - \underline{\hat{\sigma}}_a \Sigma_a(\underline{r}, 0)] \int_{4\pi} \psi(\underline{r}, \underline{\Omega}', t) d\underline{\Omega}' + \frac{1}{4\pi} Q(\underline{r}, t) \quad (12)$$

and Eqs. (7) as

$$\frac{\partial}{\partial t} \hat{N}(\underline{r}, t) = \frac{1}{\langle N(\underline{r}) \rangle} \hat{A} \hat{N}(\underline{r}, t) \Sigma_a(\underline{r}, 0) \int_{4\pi} \psi(\underline{r}, \underline{\Omega}, t) d\underline{\Omega} + \underline{\Lambda} \hat{N}(\underline{r}, t) , \quad (13)$$

where the elements of the dimensionless absorption matrix \hat{A} are given by

$$\hat{a}_{ij} = \begin{cases} -\sigma_a^j / \langle \sigma_a \rangle , & i = j . \\ f_{ij} \sigma_a^j / \langle \sigma_a \rangle , & i \neq j . \end{cases} \quad (14)$$

The system of equations given by Eqs. (12) and (13) is now in a form to which we can directly apply the classic asymptotic diffusion limit scaling of Larsen and Keller [1, 14]. We first introduce the asymptotic diffusion limit scaling, with ϵ a formal small parameter $0 < \epsilon \ll 1$, into Eq. (12):

$$\begin{aligned} \frac{\epsilon}{v} \frac{\partial}{\partial t} \psi(\underline{r}, \underline{\Omega}, t) + \underline{\Omega} \cdot \underline{\nabla} \psi(\underline{r}, \underline{\Omega}, t) + \hat{N}^T(\underline{r}, t) \hat{\sigma}_t \frac{\Sigma_t(\underline{r}, 0)}{\epsilon} \psi(\underline{r}, \underline{\Omega}, t) = \\ \frac{1}{4\pi} \hat{N}^T(\underline{r}, t) \left[\hat{\sigma}_t \frac{\Sigma_t(\underline{r}, 0)}{\epsilon} - \hat{\sigma}_a \epsilon \Sigma_a(\underline{r}, 0) \right] \int_{4\pi} \psi(\underline{r}, \underline{\Omega}', t) d\underline{\Omega}' + \frac{1}{4\pi} \epsilon Q(\underline{r}, t) , \end{aligned} \quad (15)$$

The small dimensionless parameter ϵ can be physically interpreted as the ratio of an initial typical neutron mean free path to a typical dimension of the spatial domain. Therefore, Eq. (15) describes a system that is initially optically thick with small, of $O(\epsilon)$, absorption and sources and weak, of $O(\epsilon)$, temporal variation of the angular flux. The further spatial and temporal evolution of the material properties is determined by the function $\hat{N}(\underline{r}, t)$. We next assume that the radioactive decay constants are small, of $O(\epsilon)$, and introduce the asymptotic diffusion limit scaling into Eqs. (13) to obtain

$$\epsilon \frac{\partial}{\partial t} \hat{N}(\underline{r}, t) = \frac{1}{\langle N(\underline{r}) \rangle} \hat{A} \hat{N}(\underline{r}, t) \epsilon \Sigma_a(\underline{r}, 0) \int_{4\pi} \psi(\underline{r}, \underline{\Omega}, t) d\underline{\Omega} + \epsilon \underline{\Lambda} \hat{N}(\underline{r}, t) . \quad (16)$$

Eqs. (16) describe a system with weak, of $O(\epsilon)$, temporal variation of the isotopic number densities as a result of neutron absorption and radioactive decay.

We assume that the neutron angular flux $\psi(\underline{r}, \underline{\Omega}, t)$ and the dimensionless isotopic number densities $\hat{N}(\underline{r}, t)$ have the following power series expansions in the small parameter ϵ :

$$\psi(\underline{r}, \underline{\Omega}, t) = \sum_{n=0}^{\infty} \epsilon^n \psi_n(\underline{r}, \underline{\Omega}, t) , \quad (17)$$

$$\hat{N}(\underline{r}, t) = \sum_{n=0}^{\infty} \epsilon^n \hat{N}_n(\underline{r}, t) . \quad (18)$$

To proceed, we introduce Eqs. (17) and (18) into Eq. (15) and equate coefficients of like powers of ϵ . Collecting the coefficients of $O(\epsilon^0)$ and dividing by common multipliers gives

$$\psi_0(\underline{r}, \underline{\Omega}, t) = \frac{1}{4\pi} \int_{4\pi} \psi_0(\underline{r}, \underline{\Omega}', t) d\underline{\Omega}' . \quad (19)$$

Eq. (19) has the isotropic solution

$$\psi_0(\underline{r}, \underline{\Omega}, t) = \frac{1}{4\pi} \phi_0(\underline{r}, t) , \quad (20)$$

where the function $\phi_0(\underline{r}, t)$ is currently unspecified. Therefore, the solution of the neutron transport problem specified by Eq. (15) is isotropic to leading order. The goal of the remainder of the analysis is to determine the equation that the function $\phi_0(\underline{r}, t)$ satisfies. Collecting the coefficients of $O(\epsilon^1)$ and using Eqs. (19) and (20) to cancel terms, we obtain

$$\psi_1(\underline{r}, \underline{\Omega}, t) = -\frac{1}{4\pi} \left[\hat{N}_0^T(\underline{r}, t) \hat{\sigma}_t \Sigma_t(\underline{r}, 0) \right]^{-1} \underline{\Omega} \cdot \nabla \phi_0(\underline{r}, t) + \frac{1}{4\pi} \int_{4\pi} \psi_1(\underline{r}, \underline{\Omega}', t) d\underline{\Omega}' . \quad (21)$$

Collecting the coefficients of $O(\epsilon^2)$, we obtain

$$\begin{aligned} \frac{1}{v} \frac{\partial}{\partial t} \psi_0(\underline{r}, \underline{\Omega}, t) + \underline{\Omega} \cdot \nabla \psi_1(\underline{r}, \underline{\Omega}, t) + \hat{N}_0^T(\underline{r}, t) \hat{\sigma}_t \Sigma_t(\underline{r}, 0) \psi_2(\underline{r}, \underline{\Omega}, t) \\ + \hat{N}_1^T(\underline{r}, t) \hat{\sigma}_t \Sigma_t(\underline{r}, 0) \psi_1(\underline{r}, \underline{\Omega}, t) = \frac{1}{4\pi} \hat{N}_0^T(\underline{r}, t) \hat{\sigma}_t \Sigma_t(\underline{r}, 0) \int_{4\pi} \psi_2(\underline{r}, \underline{\Omega}', t) d\underline{\Omega}' \\ + \frac{1}{4\pi} \hat{N}_1^T(\underline{r}, t) \hat{\sigma}_t \Sigma_t(\underline{r}, 0) \int_{4\pi} \psi_1(\underline{r}, \underline{\Omega}', t) d\underline{\Omega}' \\ - \frac{1}{4\pi} \hat{N}_0^T(\underline{r}, t) \hat{\sigma}_a \Sigma_a(\underline{r}, 0) \int_{4\pi} \psi_0(\underline{r}, \underline{\Omega}', t) d\underline{\Omega}' + \frac{1}{4\pi} Q(\underline{r}, t) . \end{aligned} \quad (22)$$

Inserting Eqs. (19)–(21) into Eq. (22), operating by $\int_{4\pi} (\cdot) d\underline{\Omega}$, and cancelling like terms, we obtain

$$\begin{aligned} \frac{1}{v} \frac{\partial}{\partial t} \phi_0(\underline{r}, t) - \frac{1}{3} \nabla \cdot \left[\hat{N}_0^T(\underline{r}, t) \hat{\sigma}_t \Sigma_t(\underline{r}, 0) \right]^{-1} \nabla \phi_0(\underline{r}, t) \\ + \hat{N}_0^T(\underline{r}, t) \hat{\sigma}_a \Sigma_a(\underline{r}, 0) \phi_0(\underline{r}, t) = Q(\underline{r}, t) . \end{aligned} \quad (23)$$

Next we introduce Eqs. (17) and (18) into Eqs. (16) and equate coefficients of like powers of ϵ . Collecting the coefficients of $O(\epsilon^0)$ and using Eq. (20) gives

$$\frac{\partial}{\partial t} \hat{N}_0(\underline{r}, t) = \frac{1}{\langle N(\underline{r}) \rangle} \hat{A} \hat{N}_0(\underline{r}, t) \Sigma_a(\underline{r}, 0) \phi_0(\underline{r}, t) + \underline{\Lambda} \hat{N}_0(\underline{r}, t) . \quad (24)$$

Returning to dimensional quantities in Eqs. (23) and (24), we finally obtain

$$\frac{1}{v} \frac{\partial}{\partial t} \phi_0(\underline{r}, t) - \nabla \cdot \frac{1}{3 \hat{N}_0^T(\underline{r}, t) \hat{\sigma}_t} \nabla \phi_0(\underline{r}, t) + \hat{N}_0^T(\underline{r}, t) \underline{\sigma}_a \phi_0(\underline{r}, t) = Q(\underline{r}, t) , \quad (25)$$

and

$$\frac{\partial}{\partial t} N_0(\underline{r}, t) = \underline{A} N_0(\underline{r}, t) \phi_0(\underline{r}, t) + \underline{\Lambda} N_0(\underline{r}, t) . \quad (26)$$

Eq. (25) is the standard time-dependent neutron diffusion equation coupled with Eqs. (26), the Bateman equations, describing the temporal evolution of the isotopic number densities as a result of isotopic depletion and radioactive decay. The asymptotic analysis demonstrates that the standard model for reactor analysis consisting of the neutron diffusion equation coupled with the Bateman equations is valid for the situation of an initially optically-thick system with weak absorption, sources, and temporal derivatives of the angular flux. In addition, the temporal variation of the isotope number densities as a result of absorption and radioactive decay must be small.

3. CONCLUSIONS

In this paper, we have described an asymptotic analysis of the coupled nonlinear system of equations describing time-dependent three-dimensional monoenergetic neutron transport and isotopic depletion and radioactive decay. The classic asymptotic diffusion scaling of Larsen and Keller [1] was applied to this coupled nonlinear system of equations in a medium of specified initial isotopic composition. The analysis demonstrates that to leading order the neutron transport equation limits to the standard time-dependent neutron diffusion equation with macroscopic cross sections whose number densities are determined by the standard system of ordinary differential equations, the so-called Bateman equations, describing the temporal evolution of the nuclide number densities. For this asymptotic limit to hold, the temporal evolution of the isotopic number densities as a result of radioactive decay must be weak.

In future work, we plan to continue the analysis presented in this paper to include asymptotic analyses of both the initial and the boundary layers. Because most realistic problems include energy dependence through the multigroup approximation, we also hope to extend our analysis to multigroup neutron transport problems.

ACKNOWLEDGEMENTS

This work was performed under the auspices of the U.S. Department of Energy by the University of California Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

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